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(54) Cavity-locked ring down spectroscopy

(57) A cavity ring-down spectroscopy system comprising a cavity for holding a sample, a light source for generating locking light and sampling light of different polarization and/or of different frequencies, locking means for locking the cavity and the light source, and detection means for detecting sampling light and for allowing determination of ring-down rates indicative of an absorption of the sampling light by the sample.

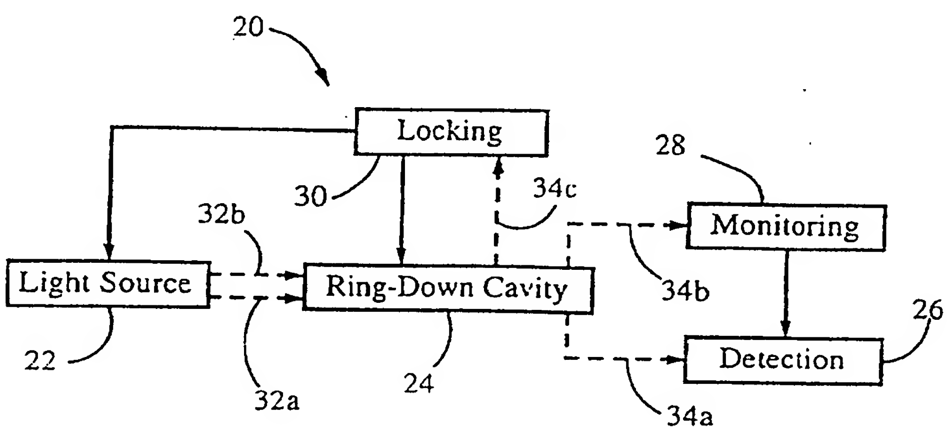

FIG. 1-A

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Description

[0001] This invention relates to cavity ring-down spectroscopy (CRDS), and in particular to systems and methods for locking a laser and a ring-down cavity for CRDS.

[0002] Traditional spectroscopic methods are limited in sensitivity to approximately one part per ten thousand ($1:10^4$) to one part per hundred thousand ($1:10^5$), whereas cavity ring-down spectroscopy (CRDS) allows making absorption measurements with sensitivities on the order of one part per ten million ($1:10^7$) to one part per billion ($1:10^9$) or higher.

[0003] In a conventional CRDS system, the sample (absorbing material) is placed in a high-finesse stable linear optical resonator. The intensity of a light pulse introduced in the resonator decreases in time. For an empty cavity, the intracavity light intensity follows an exponential decay characterised by a ring-down rate that depends only on the reflectivity of the mirrors, the separation between the mirrors, and the speed of light in the cavity. If a sample is placed in the resonator, the ring-down is accelerated. Under suitable conditions, the light decay remains exponential. An absorption spectrum for the sample is obtained by plotting the reciprocal of the ring-down rate versus the wavelength of the incident light.

[0004] Conventional pulsed CRDS systems provide a number of challenges. In systems using pulsed lasers, data acquisition rates may be limited by the repetition rate of the pulse laser source. Moreover, the intensity of the light coupled into an out of the cavity may be small, as a consequence of the relatively low spectral overlap between cavity modes and laser linewidth, as well as a lack of significant light buildup within the cavity.

[0005] US Patent No. 5,528,040 (Lehmann) discloses the use of continuous-wave (c.w.) laser sources for CRDS, in particular laser diode (LD) sources using controlled optical feedback from a reference cavity. Such optical feedback may, however, cause instability in the laser operation. Moreover, locking was turned off during measurements, such that each measurement required re-locking the laser and cavity, which may limit the repetition of rates achievable with the system.

[0006] As least in certain embodiments the present invention is able to provide a system and method for stable and reliable locking of a laser and a ring-down cavity for CRDS, a CRDS system allowing continuous locking of the laser and ring-down cavity over multiple rind-down measurements, a CRDS system allowing high measurement repetition rates, a CRDS system in which locking and sampling light have different frequencies, such that the locking light does not affect measurement values obtained using the sampling light, a CRDS system with reduced shot-to-shot variation in the ring-down constant, a CRDS system with reduced baseline noise, a CRDS system allowing locking of the laser and the ring-down cavity without requiring an external reference cavity, a CRDS system allowing reduced optical feedback from the ring-down cavity to the laser, and a system allowing relatively simply coupling of locking light extending from the CRDS cavity to locking components.

[0007] Thus, the present invention provides a cavity ring-down spectroscopy system comprising:

(a) a ring-down resonant cavity for holding a sample;
(b) a light source comprising a laser for generating locking light and sampling light incident on the cavity, the locking light and sampling light being of different polarization and/or of different frequency;
(c) locking means in optical communication with the cavity for locking the cavity and the light source using locking light reflected by the cavity; and
(d) detection means in optical communication with the cavity for detecting sampling light extending from the cavity and for allowing determination of ring-down rates indicative of an absorption of the sampling light by the sample.

[0008] The system of the present invention therefore uses distinct sampling and locking light beams. The sampling light is used for performing ring-down measurements, while the locking light is used for frequency-locking a ring-down resonant cavity and a c.w. or other laser. The locking light will in general be maintained continuously on while the sampling light will in general be turned on and off to allow one to obtain ring-down measurements. Continuous locking allows higher measurement repetition rates, and consequently lower noise levels. The locking and sampling light may have different frequencies such that they are effectively decoupled within the cavity, ensuring that the locking light does not affect ring-down measurements performed with the sampling light.

[0009] In a preferred embodiment, the ring-down cavity is a ring-shaped cavity. For a given mode, the ring cavity has distinct resonant frequencies and mirror losses for s-polarized and p-polarized light. If intracavity losses are higher for p-polarized light than for s-polarized light, the sampling light is preferably s-polarized with respect to the cavity, while the locking light is p-polarized. The use of a ring-shaped cavity provides for a desirable separation between the resonant frequencies of a given mode, allowing tuning of each of the sampling and locking light to the same cavity mode without coupling the sampling and locking light. The use of a ring-shaped cavity provides the further advantage of allowing a reduction in optical feedback to the laser.

[0010] At least one of the locking and sampling light is preferably frequency-shifted to ensure that the sampling and locking light are decoupled within the cavity. Preferably, an acousto-optic modulator (AOM) is positioned in the path of the sampling light. The AOM is used to introduce a difference between the frequencies of the sampling and locking light. The AOM is also used as a

switch for turning the sampling light on and off to allow the performance of ring-down measurements.

[0011] In alternative embodiments, the ring-down cavity may be linear or folded. The sampling and locking light can then be tuned to the same transverse mode (e. g. TEM_{00}) corresponding to adjacent longitudinal modes. The frequency-shift between the sampling and locking light is then equal to one free spectral range of the cavity.

[0012] The invention is further illustrated with reference to the accompanying drawings, in which:

Fig. **1-A** is a schematic high-level diagram of a preferred system of the present invention,

Fig. **1-B** is a schematic diagram of the optical components of the system of Fig. 1-**A,**

Fig. **1-C** illustrates the locking components of the system of Fig. **1-A,**

Fig. **2** shows that locking components in an alternative embodiment of the present invention,

Fig. **3** shows an alternative embodiment of the present invention, using a linear ring-down cavity,

Fig. **4** shows scans of the cavity frequency response for p-polarized light and s-polarized light for an exemplary system of the present invention,

Fig. **5** shows a measured plot of the spectral density of the frequency difference fluctuation as a function of frequency difference for the exemplary system characterised by Fig **4**, and

Fig. **6** shows a spectrum of water vapour in ambient room air recorded using the exemplary system characterised by Fig **4**.

[0013] The following description illustrates the invention by way of example. In the following discussion, unless otherwise stated, polarization directions are to be considered relative to the ring-down resonant cavity of the system. Unless otherwise stated, the term "ring-down" encompass each of decay and build-up (sometimes referred to as "ring-up") of light withing the resonant cavity.

[0014] Fig. **1-A** is a schematic high-level diagram of a preferred system **20** of the present invention. Optical connections are illustrated by dashed lines, while electrical connections are illustrated by solid lines. System **20** comprises a tunable monochromatic light source **22**, a ring-down resonant cavity **24** in optical communication with light source **22**, as well as sampling detection electronics **26**, monitoring (diagnostics) electronics **28** and locking electronics **30** all in optical communication with cavity **24**. Locking electronics **30** are electrically connected to light source **22** and cavity **24**, while monitoring electronics **28** are electrically connected to sampling detection electronics **26**.

[0015] Cavity **24** holds a sample of interest in an intracavity light path. Light source **22** generates c.w. input sampling light **32a** and locking light **32b** incident on cavity **24**. Input sampling light **32a** has a wavelength within an absorption region of interest of the sample. Input locking light **32b** has a predetermined wavelength relationship with sampling light **32a**. Sampling light **32a** and locking light **32b** have different frequencies within cavity **24**. Preferably, sampling light **32a** is s-polarized while locking light **32b** is p-polarized.

[0016] Output light **34a-c** extending from cavity **24** is incident on detection electronics **26**, monitoring electronics **28,** and locking electronics **30**, respectively. Output light **34a-c** comprises output sampling light **34a** incident on sampling detection electronics **26**, output monitoring light **34b** incident on monitoring electronics **28,** and output locking light **34c** incident on locking electronics **30**. Locking electronics **30** frequency-lock light source **22** and cavity **24**, ensuring that the linewidth of intracavity sampling light overlaps the linewidth of a mode of interest of cavity **24**. Sampling detection electronics **26** determine ring-down rates, absorption spectra, and species quantities for the sample of interest. Monitoring electronics **28** monitor the intensity and transverse mode structure of intracavity light. The intensity of intracavity light is used to characterize the locking of light source **22** and cavity **24.** In one embodiment, measurements of peak intracavity light intensity are used to adjust the frequency separation between locking and sampling light.

[0017] Fig. 1-B illustrates the optics of system **20.** Locking electronics 30 are illustrated in Fig. **1-C.** For clarity of presentation, various standard elements such as lenses and mirrors used for focusing and directing beams are not described in detail; such elements are well known in the art.

[0018] Referring to Fig. **1-B,** light source **22** comprises a continuous wave (c.w.) laser **40** for generating primary light **42**, and optical components for generating input sampling light **32a** and locking light **32b** from primary light **42**. Laser **40** preferably has a linewidth on the order of 1 MHz and a useful output power of at least 100 µW. Laser **40** is preferably, a tunable external-cavity diode laser (ECDL). Other suitable light sources include solid state and dye lasers, as well as C.W. optical parametric oscillators (OPOs). Laser **40,** including its controller, is mounted within a Faraday cage (not shown), for shielding laser **40** from stray electromagnetic interference which may otherwise cause instability in the temperature controller of laser **40**.

[0019] An optical isolator **44** is positioned in the path

of primary light **42**, in optical communication with laser **40**. Isolator **44** reduces the optical feedback caused by back reflections into laser **40**, and controls the polarization of primary light **42**. Isolator **44** is preferably a Faraday isolator including a half-wave plate adjusted to generate p-polarized light. A beam expander **46** and a beam splitter **48** are positioned in sequence in front of isolator **44**, so as to receive light extending from isolator **44**. Primary light **42** passes through beam expander **46** before being split by beam splitter **48** into sampling light **32a** and locking light **32b**.

[0020] An acousto-optic modulator (AOM) **54** is positioned in the path of sampling light **32a**, between laser **40** and cavity **24**. AOM **54** is driven by an RF signal generated by a voltage-controlled oscillator (not shown). AOM **54** is flanked by focusing and mode-matching optics **56a-b**. Optics **56a** comprise a mode-matching lens for selecting TEM_{00} light for passage to cavity **24**, and a focusing lens for collimating sampling light **32a** onto AOM **54**. Optics **56b** comprise lenses for recollimating sampling light **32a** after passage through AOM **54**. A half-wave plate **57** is positioned in the path of sampling light **32a**, for changing its polarization state to s-polarized. In the arrangement shown, AOM **54** is optimized for modulating p-polarized light, and thus half-wave plate **57** is positioned after AOM **54**.

[0021] AOM **54** acts as a switch, for modulating (e.g. switching on and off) the intensity of sampling light **32a** reaching cavity **24** so as to allow sampling light **32a** to ring down within cavity **24**.

[0022] AOM **54** may be used to generate pulses or continuous wave step inputs, among others. AOM **54** also acts as a frequency-shifter, for shifting the frequency of sampling light **32a** incident on cavity **24** such that s-polarized sampling light **32a** is tuned to a desired mode of cavity **24**. AOM **54** shifts the frequency of sampling light **32a** by an amount equal to the frequency of the RF signal driving AOM **54**. Preferably, the first order beam from AOM **54** is directed to cavity **24**. An aperture **50** is positioned in the optical path between AOM **54** and cavity **24**, to select the first order beam from AOM **54** for passage to cavity **24** while blocking the zero order beam.

[0023] A phase-modulator (PM) **58** is situated in the path of locking light **32b**, between laser **40** and cavity **24**. PM **58** is preferably an electro-optic resonant phase modulator. In the particular arrangement shown, PM **58** is optimized for modulating s-polarized light, and is thus flanked by half-wave plates **60a-b**. Half-wave plates **60a-b** control the polarization of light within PM **58** so as to maximize the electro-optic effect within PM **58** for the particular crystal orientation of PM **58**. PM **58** introduces locking sidebands into locking light **32b**. The sidebands are separated from the central frequency ω_L of locking light **32b** by a predetermined frequency separation ω_f larger than the linewidths of laser **40** and the mode of interest of cavity **24**. The sideband at ω_L-ω_f is phase-shifted by π (180°) with respect to the sideband at ω_L+ω_f. A mode-matching lens **62** is situated in the path of locking light **32b**, for selecting the TEM_{00} mode for passage to cavity **24**.

[0024] A polarizing beam splitter **64** is positioned in the paths of sampling light **32a** and locking light **32b**, facing cavity **24**. Beam splitter **64** combines and directs sampling light **32a** and locking light **32b** toward cavity **24**. Beam splitter **64**, isolator **44**, and half-wave plates **57, 60a-b** act as polarization control elements, selecting s-polarized sampling light **32a** and p-polarized locking light **32b** for passage to cavity **24**.

[0025] Cavity **24** comprises an input mirror **66a**, an intermediate mirror **66b**, and an output mirror **66c**. Mirrors **66a-c** define a closed, triangular intracavity light path **68**. A gas sample **72** is situated within light path **68**, and fills cavity **24**. Sample **72** is an optically passive material (i.e. not a gain medium). Input mirror **66a** and output mirror **66c** are flat moderately-reflective mirrors. Intermediate mirror **66b** is a highly-reflective spherical mirror. Intermediate mirror **66b** is mounted on a piezoelectric stage **74**. Stage **74** is used as a cavity pathlength control element. Stage **74** controls the position of mirror **66b** relative to mirrors **66a** and **66c**, thus controlling the pathlength of cavity **24** and the resonant frequency of any given mode of cavity **24**.

[0026] Preferably, the thermal expansion and stress tensor coefficients of cavity **24** are optimized so as to minimize cavity deformations (due to stretching, bowing, bending, etc.) and mechanical resonances. Therefore, it is preferred that the optical elements of cavity **24** are monolithically integrated. Mirrors **66a,c** and stage **74** are rigidly attached to a common thermally stabilized block of material (not shown), preferably made of a low-thermal-expansion glass such as mica ceramic. The stable, monolithic attachment of mirrors **66a,c** and stage **74** serves to reduce changes in the shape of cavity **24** due to mechanical vibrations and/or temperature variations. Such changes in shape could cause drift in the mode frequencies of cavity **24**.

[0027] Input light **32a-b** enters cavity **24** through input mirror **66a** and generates an intracavity traveling wave along light path **68**. Input light **32a-b** is not normal to mirror **66a**, such that light reflected by mirror **66a** is not colinear with light incident on mirror **66a**. The finesse of cavity **24** is high enough that the intracavity traveling wave propagates around cavity **24** multiple times. Intracavity locking (p-polarized) and sampling (s-polarized) light encounter different losses within cavity **24**, and have different resonant frequencies for a given longitudinal mode of cavity **24**. Detection light **34a** and monitoring light 34b exit cavity **24** through output mirror **66c**.

[0028] A polarizing beam splitter **76** is in optical communication with output mirror **66c**. Beam splitter **76** is positioned in an optical path between mirror **66c** and a sampling detector **82**. Beam splitter **76** directs s-polarized sampling light **34a** to sampling detector **82**. Detector **82** is preferably a photodiode as described by Harb

et al. in *Phys. Rev. A.* 54:4370 (1996), optimized to have a high gain. A polarization separator **78** and a focusing lens **80** are positioned in the optical path between beam splitter **76** and detector **82.** Suitable polarization separators include Brewster windows, Glan-Taylor, Glan-Thomson, or Wollaston prisms, or Thomson beamsplitters, among others. Polarization separator **78** selects s-polarized sampling light **34a** for transmission to sampling detector **82**.

[0029] Ring-down electronics **90** are in electrical communication with sampling detector **82.** Ring-down electronics **90** determine ring-down rates and corresponding absorption spectra for sample **72**, by analyzing ring-down waveforms detected by sampling detector **82.** Ring-down electronics **90** may also determine quantities of trace species of interest in sample **72.** The intensity of output sampling light **34a** incident on detector **82** is indicative of the intensity of sampling light within cavity **24**, and thus of the interaction of sample **72** with intracavity sampling light. Ring-down electronics **90** are conventional. Ring-down electronics **90** may be implemented for example using computer software in an experimental setting, or using dedicated hardware in a process control device in a manufacturing setting.

[0030] Monitoring detectors **86, 88** are in optical communication with mirror **66c.** Beam-splitter **76** directs p-polarized monitoring light **34b** to monitoring detectors **86, 88**, through a beam splitter **92** and a focusing lens **94**. Detector **86** is preferably a photodiode as described in the above-referenced article by Harb et al. Detector **86** is used to monitor the power of intracavity light. Detector **86** may be connected through monitoring servo electronics (not shown) to AOM **54**, for adjusting the driving frequency of AOM **54** to compensate for changes in the frequency difference between sampling and locking resonances. Such changes may be caused by sample dispersion.

[0031] Detector **88** is preferably a charge-coupled-device array (CCD camera). Detector **88** is used to monitor the transverse mode structure of intracavity light, ideally to ascertain that only TEM_{00} light propagates within cavity **24.** Monitoring processing electronics **96** are in electrical communication with detectors **86** and **88**, and with ring-down electronics **90.** Electronics **96** communicate recorded monitoring data to ring-down electronics **90** for storage with corresponding ring-down data.

[0032] A locking detector **100** is in optical communication with input mirror **66a**, and is positioned to capture output locking light **34c** extending from input mirror **66a.** Locking detector **100** is preferably a photodiode similar to that used for monitoring detector **86**, optimized for high bandwidth in order to render the locking sidebands with minimal distortion and noise. A polarization separator **102** and a focusing lens **104** are positioned in an optical path of locking light **34c**, between mirror **66a** and locking detector **100.** Light extending from input mirror **66a** consists of a reflected component reflected by input mirror **66a**, and of a leakage component transmitted through input mirror **66a** from cavity **24**. Polarization separator **102** selects p-polarized locking light **34c** for transmission to detector **100.**

[0033] Detector **100** is part of locking electronics **30.** Fig. **1-C** shows a schematic diagram of locking electronics **30.** The design of locking electronics **30** is based on the Pound-Drever locking system. For detailed information on Pound-Drever locking see the article by Drever et al. in *Appl. Phys.* B 31:97-105 (1983), herein incorporated by reference.

[0034] Referring to Fig. **1-C**, a high-pass filter (HPF) **108** is electrically connected to the output of detector **100.** The pass-threshold of HPF **108** is less than the frequency separation ω_f between the central frequency and the locking sidebands of the signal detected by detector **100.** A band-pass filter (BPF) **110** is connected to the output of HPF **108.** The passband of BPF **110** is centered at ω_f. The output of BPF **110** is connected to an input of a mixer **112.** Another input of mixer **111** is connected to a first output of a voltage-controlled oscillator (VCO) **106** capable of generating a driving signal at the frequency ω_f. A second output of VCO **106** is connected to PM **58**, for driving PM **58.** An output of mixer **112** is connected to a low-pass filter (LPF) **114.** The pass-threshold of LPF **114** is less than $2\omega_f$. The output of LPF **114** is connected to servo electronics **116**, which are in turn connected through an amplifier **118** to stage **74.**

[0035] VCO **106** generates a driving signal at ω_f for driving PM **58.** PM **58** inserts locking sidebands into locking light **32b** at frequencies $\omega_L \pm \omega_f$, where ω_L is the frequency of the central band of locking light **32b.** Output locking light **34c** is formed by the reflection of locking light **32b** and the locking light leaking from cavity **24.** The amplitudes of the sidebands of locking light **34c** depend on the tuning of each sideband to cavity **24**, and consequently on the difference between ω_L and the corresponding resonant frequency of a given mode of interest of cavity **24.** For example, if ω_L is higher than a corresponding resonant frequency of cavity **24**, the reflected sideband at $\omega_L - \omega_f$ has a lower intensity than the reflected sideband at $\omega_L + \omega_f$.

[0036] Locking light **34c** detected by detector **100** includes a frequency component at ω_L and a frequency component at ω_f. Typically, ω_L is on the order of 10^{14} Hz, while ω_f is on the order of 10^7 Hz. Detector **100** is not fast-enough to time-resolve the ω_L component of locking light **34c.** Consequently, the electric signal generated by detector **100** includes a component centered at zero frequency, corresponding to the ω_L frequency component of locking light **34c.** Detector **100** can time-resolve the ω_f component of locking light **34c**, and thus the generated electric signal includes a component centered at ω_f. HPF **108** and BPF **110** select the ω_f frequency component for passage to mixer **112.**

[0037] Mixer **112** mixes the signals received from VCO **106** and detector **100** to generate an error signal centered at zero frequency. Mixer **112** effectively differences the sideband signals to generate the error signal.

The sign of the error signal indicates whether ω_L is higher or lower than the corresponding resonant frequency of cavity **24**. LPF **114** eliminates higher-harmonics (e.g. $2\omega_f$) from the signal generated by mixer **112**, selecting the zero-frequency error signal for passage to servo electronics **116**. Servo electronics **116** translate the error signal into an appropriate motion of stage **74**, for adjusting the pathlength and thus resonant frequency of interest of cavity **24** to ensure that a desired mode of cavity **24** is tuned to ω_L.

[0038] Locking light **32b** is continuously turned on during the operation of system **20**, for ensuring continuous locking of cavity **24** and light source **22**. Sampling light **32a** is selectively turned off after sufficient accumulation of light within cavity **24** during periods of c.w. operation, to allow measurements of the ring-down of sampling light **32a** within cavity **24**. Maintaining locking light **32b** turned on while performing multiple ring-down measurements allows relatively high repetition rates (e.g. kHz to tens of kHz) for system **20**.

[0039] Fig. **2** is a simplified diagram illustrating an alternative system **20'** according to the present invention. Locking electronics **30'** control the frequency of light emitted by a laser **40'**, so as to tune the central band of locking light **32b** to cavity **24**. The output of mixer **112** is connected through LPF **114**, servo electronics **116'**, and an amplifier **118'** to a frequency controller of laser **40'**. If ω_L is higher than the resonant frequency of interest of cavity **24**, servo electronics **116'** control laser **40'** to reduce its emission frequency so as to tune ω_L to the desired mode of cavity **24**. Similarly, ω_L is increased if it is lower than desired.

[0040] Fig. 3 is a simplified diagram illustrating another alternative system **20"** of the present invention. System **20"** includes a linear ring-down cavity **24"**. A polarizing beam splitter **65** and a quarter-wave plate **67** are positioned in sequence between PBS **64** and cavity **24"**, in the optical path of the sampling and locking light. PBS **65** directs incident locking and sampling light toward cavity **24"**, while directing reflected locking light toward detector **100**. The frequencies of the sampling and locking light are tuned to the same transverse mode (e.g. TEM_{00}) but different (e.g. adjacent) longitudinal modes of cavity **24"**. The frequency difference between the sampling and locking light is equal to one free spectral range (FSR) of cavity **24"**. For typical FSR values of hundreds of MHz, a conventional AOM is well suited for introducing the desired frequency difference between the sampling and locking light.

[0041] Baseline noise in CRDS is caused in large part by concurrent excitation and beating of multiple modes within the ring-down cavity (RDC), or by consecutive excitation of different modes. Different transverse/longitudinal modes have different resonant frequencies, and ring-down at different rates in an empty cavity. Ensuring that a single mode is excited within the ring-down cavity allows a significant improvement in system signal-to-noise ratios.

[0042] Because frequency fluctuations in the laser source appear identically in both sampling and locking light, if a locking light mode is locked to the cavity, then a corresponding sampling light mode is also locked to the cavity. The AOM used for switching the sampling light on and off provides the appropriate frequency shift to the sampling light, so that it can couple into the same transverse mode as the locking light.

[0043] Using a ring-shaped ring-down cavity in a system of the present invention provides a number of advantages. The use of a ring-shaped cavity provides for the desired separation between the resonant frequencies for sampling and locking light of different polarizations, allows a simplification in the optics used for coupling locking light into and out of the cavity, and allows a reduction in optical feedback to the laser.

[0044] It is generally well-known from the physics of optical interfaces that non-normal incidence of linearly polarized light on a dielectric interface will usually result in different responses for p-polarized light (PPL) and s-polarized light (SPL). Thus, an optical ring resonator constructed with a geometry using non-normal incidence reflectors (e.g., an isosceles triangle) will actually consist of two superimposed nondegenerate Fabry-Perot cavities: a p-polarization cavity (PPC), and an s-polarization cavity (SPC).

[0045] Non-normal incidence dielectric mirrors typically have a lower reflectivity for PPL than they do for SPL. Consequently, a ring resonator will usually consist of a reduced finesse PPC, and a higher finesse SPC. The ring resonator PPC and SPC transverse mode structures are identical, because they are determined only by geometrical considerations. The unequal phase shifts accrued during non-normal reflection of PPL and SPL will result in a fixed frequency difference between the PPC and SPC frequency responses. PPC and SPC free spectral ranges differ by the contribution of the derivative of the mirror phase shift, which was estimated to be a few tens of kHz.

[0046] Because orthogonal polarizations are easily separated with polarizing optics, the simultaneous use of PPL and SPL provides a straightforward solution for separating the cavity locking problem from the actual ring-down measurement. The use of the lower finesse PPC for locking improves the feedback locking signal, and relaxes requirements on servo bandwidth, feedback gain, and differential sensitivity to the frequency difference between the laser and cavity. Absolute wavelength accuracy at each point is determined by the PPC finesse and locking servo quality, rather than by laser linewidth or cavity mode sweep range. The improved frequency stability enables a substantial increase in the resolution achievable with such a system.

[0047] The use of a ring cavity also allows a significant simplification in the optics used to couple locking light into locking components. In a system using a linear or folded cavity, the optical signal extending from the cavity input is colinear with the light beam incident on the cavity

input. As a result, optical components such as half-wave plates, analyzers, and beam-splitters may be needed to selectively couple the locking light reflected by the cavity input into the locking electronics. Furthermore, ring cavities do not reflect light directly back into the light source, so that optical feedback problems are greatly reduced.

[0048] The following example is intended to illustrate a particular implementation of the invention, and should not be construed to limit the invention.

Example

[0049] A system of the present invention was used to generate spectra of water vapor in ambient air. The laser was a commercial ECDL (New Focus: 6226-H032), tunable from 833.2 nm to 862.5 nm. The laser output varied from about 9 to 13 nW for 60 mA drive current, over the entire tuning range. A 25-35 dB Faraday isolator (New Focus: 5568) was used to reduce residual back reflections into the laser. The input and output mirrors of the RDC (CVI:TLM2-800-45S-1037) had 99.93% reflectivity for SPL at 833 nm, and 99.3% reflectivity for PPL at 833 nm. The intermediate RDC mirror (REO: run C628, 7.75 mm, 840-880 nm, 1 degree wedge) had a 99.95% reflectivity at 833 nm for both polarizations.

[0050] One quarter of the primary light generated by the laser was used for locking and three quarters for sampling. The input sampling light was focused to a spot size of 60 µm at the center of the AOM (Brimrose: GPM-400-100-960), and recollimated using 5 cm focal length lenses. The AOM was driven by a VCO with a frequency range of 300 to 535 MHz. The AOM imparted a frequency shift of 320 MHz to the input sampling light. The lenses used for TEM_{00} mode-matching of the sampling and locking light had a focal length of 127 cm. The phase modulator was a 58.5 MHz electro-optic resonant phase modulator (New Focus:4001). The fraction of laser light coupled into the TEM_{00} mode of the cavity was ~90% for PPL, and ~85% for SPL.

[0051] The VCO driving the phase-modulator was set to 58.5 MHz. A Glan-Taylor prism was used to select PPL for passage to the locking detector. The bandpass of the locking bandpass filter was centered at 60 MHz. The servo electronics consisted of a proportional integral (PI) controller coupled to a passive notch filter centered at 52 kHz (48 dB maximum attenuation, 3 dB bandwidth of 2 kHz). The notch filter was used to suppress oscillation of the piezoelectric stage at its resonant frequency of 52 kHz. The use of the notch filter improved the stability of the system and reduced its sensitivity to external perturbations such as acoustic noise. The gain of the servo electronics and associated amplifier was 60 dB, with a 3 dB bandwidth of 455 Hz.

[0052] The locking and monitoring photodiodes were optimized for a bandwidth of 90 MHz. Shot-noise-limited sideband signals were readily attainable for optical powers of hundreds of µW at the detectors. The sampling photodiode was optimized for gain, at a bandwidth of 25 MHz. The output sampling light power was on the order of µW, and the detected ring-down decay constants were on the order of hundreds of ns or higher. The monitoring photodiode received 90% of the monitoring light, while the monitoring CCD camera received the remaining 10%.

[0053] A Glan-Thomson prism was used to select SPL for passage to the sampling detector. After detection by the sampling detector, the sampling signals were amplified using a low-noise, highspeed amplifier (Stanford Research Systems:SR445), and digitized using a 10-bit, 1 GHz oscilloscope (Tektronix:11402). The decay waveforms were fitted using the Levenberg-Marquardt algorithm with the initial guess provided by a linear least squares fit of the signal logarithm, as described by Naus et al. in *ILS-XII 12th Interdisciplinary Laser* Science *Conference*. ed. A. P. Society (OSA, Rochester, New York, 1996), p. 122. All scans were performed in air at atmospheric pressure, with a water partial pressure of 12 torr.

[0054] Fig. 4 shows scans of the cavity frequency response for PPL and SPL. The zero frequency is arbitrary. The resonance frequencies for SPL and PPL were observed to be separated by 282 MHz. The resonance frequency separation changed by several tens of kHz when the cavity modes overlapped an absorption line of water. The driving frequency for the AOM was accordingly adjusted using intensity information from the monitoring electronics, to maintain an appropriate frequency separation between the locking and sampling light. For small sample quantities (e.g. for trace species detection), the frequency separation resulting from dispersion by the sample is generally small or negligible.

[0055] The discriminator slope of the error signal generated by the mixer, which converts error signal voltage into frequency difference, was used to determine fluctuations in the frequency difference between the laser frequency and the cavity resonance frequency. Fig. 5 shows a measured plot of the spectral density of the frequency difference fluctuation (noise as a function of frequency difference between the laser and cavity). The total jitter between the laser and cavity resonances was 1.1 MHz, slightly less than the free-running laser linewidth. The locking components reduced noise at low frequencies (<20 kHz). Faster servo electronics may generally be used for reducing higher frequency components of the noise.

[0056] Fig. 6 shows a spectrum of water vapour in ambient room air recorded using the system described above, and a corresponding spectrum obtained from the HITRAN96 database. The resolution of the recorded spectrum was 0.002 nm, except about the absorption peaks where the resolution was increased to 0.001 nm. Ten averaged decay waveforms (ADW) were recorded at each wavelength. Each ADW was determined by averaging 256 shots. The RMS baseline noise was 5×10^{-9} cm^{-1}, and the nominal sensitivity was 5ppm. The recorded spectrum compares favourably with the HITRAN96

spectrum in absolute frequency, linestrength, and linewidth.

[0057] The above embodiments may be altered in many ways. For example, a wide variety of wavelength ranges may be used, and the presence of a variety of species may be determined. Various locking schemes and electronics may be used.

Claims

1. A cavity ring-down spectroscopy system comprising:

   (a) a ring-down resonant cavity for holding a sample;
   (b) a light source comprising a laser for generating locking light and sampling light incident on the cavity, the locking light and sampling light being of different polarization and/or different frequencies;
   (c) locking means in optical communications with the cavity for locking the cavity and the light source using locking light reflected by the cavity; and
   (d) detection means in optical communication with the cavity for detecting sampling light extending from the cavity and for allowing determination of ring-down rates indicative of an absorption of the sampling light by the sample.

2. A system according to claim 1, wherein one of the locking light and the sampling light is s-polarized within the cavity, and the other of the locking light and the sampling light is p-polarized within the cavity.

3. A system according to claim 1 or 2, wherein the locking means is in electrical communication with at least one of the light source and the cavity, receives locking light reflected by the cavity, and locks the cavity and the light source using the locking light reflected by the cavity.

4. A system according to any preceding claim, wherein the light source further comprises polarization control optics in an optical path between the laser and the cavity, for selecting a p-polarization for the locking light, and an s-polarization for the sampling light.

5. A system according to any preceding claim, wherein the light source further comprises frequency-shifting optics in an optical path between the laser and the cavity, for frequency-shifting at least one of the locking light and the sampling light to tune said at least one of the locking and the sampling light to a desired mode of the cavity.

6. A system according to claim 5, wherein the frequency-shifting optics comprises an acousto-optic modulator positioned in an optical path of the sampling light between the laser and the cavity, for frequency-shifting the sampling light to tune the sampling light to the cavity, and for switching the sampling light to allow the sampling light to ring down within the cavity.

7. A system according to any preceding claim, wherein the cavity has a cavity pathlength control element in electrical communication with the locking means, for controlling an optical pathlength of the cavity to lock the cavity to the light source.

8. A system according to any preceding claim, wherein the light source further comprises a phase modulator in an optical path of the locking light between the laser and the cavity; and wherein the locking means comprises:

   a) a locking detector in optical communication with the cavity, for detecting the locking light reflected by the cavity;
   b) processing electronics in electrical communication with the locking detector, for generating an error signal indicative of a difference between a central frequency of the locking light and a resonant frequency of the cavity;
   c) an oscillator in electrical communication with the phase modulator, for driving the phase modulator to insert a first and a second sideband in the locking light, the first sideband being at a higher frequency and the second sideband being at a lower frequency than the central frequency; and
   d) a mixer in electrical communication with the locking detector and the oscillator, for differencing electrical signals corresponding to the first and second sidebands to generate the error signal.

9. A system according to any preceding claim in which:

   a) the laser is a continuous-wave tunable laser, in optical communication with an input to the cavity, for generating a primary light beam;
   b) the system additionally comprises beam splitting optics in an optical path between the laser and the input, for splitting the primary beam into the locking light and the sampling light incident on the input, the locking light being p-polarized and the sampling light being s-polarized within the cavity;
   c) the locking means generates an electrical feedback error signal from a reflection of the locking light from the input, and locks the laser

and the cavity using the error signal; and
d) a switch is positioned between the beam splitting optics and the input, in an optical path of the sampling beam, for switching the sampling beam to allow the sampling beam to ring down within the cavity multiple times while the laser and the cavity are locked.

10. A system according to any preceding claims, in which the detection means detects a plurality of ring-downs of sampling light extending from the cavity while the cavity and the light source are locked.

11. A system according to any preceding claim, in which the detection means detects multiple ring-downs of the sampling light extending from the cavity while the cavity and the light source are locked; the system additionally comprising ring-down means in electrical communication with the detection means, for determining ring-down rates for the ring-downs.

12. A system according to any preceding claim, wherein the cavity is a linear cavity.

13. A system according to claim 12, wherein a frequency separation between the sampling light and the locking light is substantially equal to one free spectral range of the cavity.

14. A method of locking a laser and a cavity for ring-down spectroscopy, comprising the steps of:

a) using the laser to generate sampling light and locking light incident on the cavity, wherein the sampling light and the locking light have different frequencies;
b) using the locking light to lock the cavity and the laser; and
c) obtaining a plurality of measurements of a ring-down of the sampling light while maintaining the cavity and the laser in a locked state, for generating an absorption spectrum of a sample situated within the cavity.

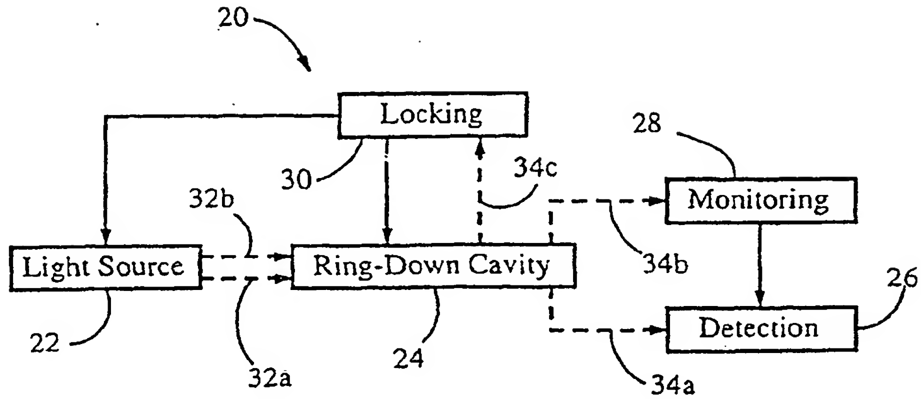

20
Locking
28
30
34c
32b
Monitoring
Light Source
Ring-Down Cavity
34b
26
Detection
22
24
32a
34a

FIG. 1-A

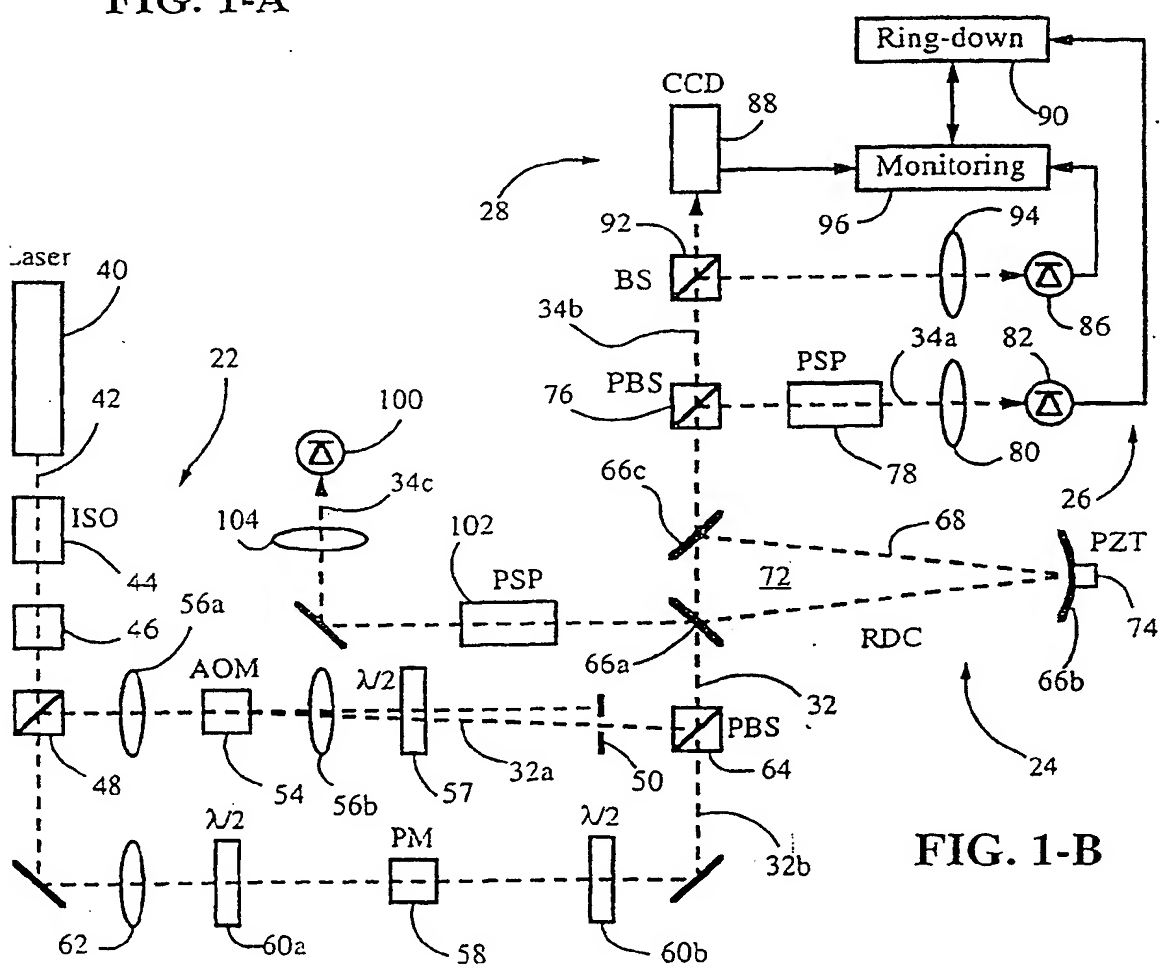

Ring-down
CCD
88
90
Monitoring
28
92
96
94
Laser
40
BS
86
34b
34a
82
22
42
76
PBS
PSP
100
80
78
34c
66c
26
ISO
104
102
68
PZT
44
PSP
72
56a
46
AOM
λ/2
66a
RDC
74
66b
32
PBS
48
54
56b
57
32a
50
64
24
λ/2
PM
λ/2
32b
62
60a
58
60b

FIG. 1-B

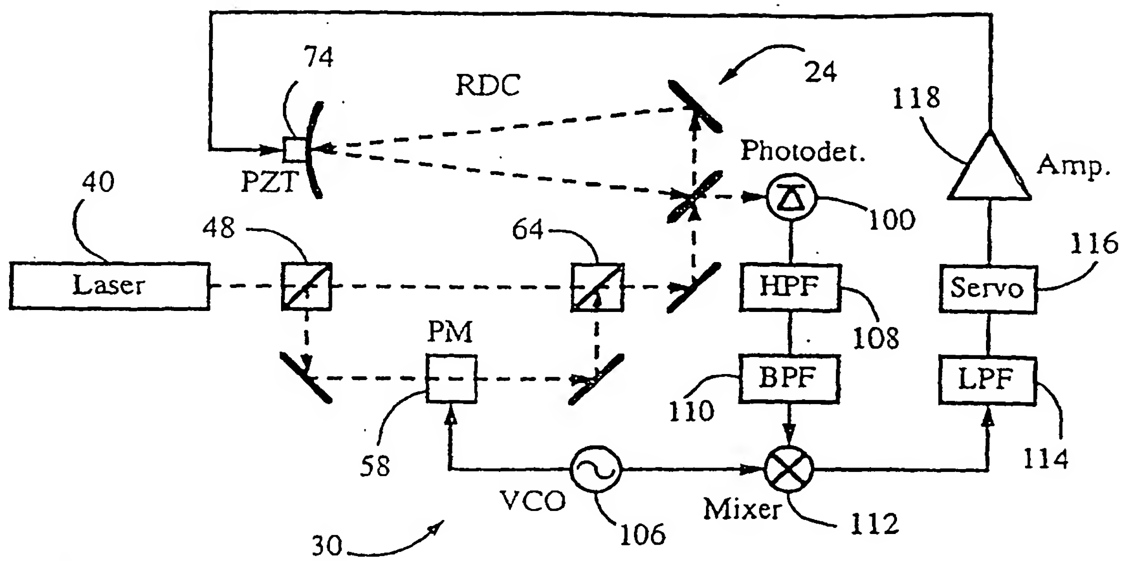

74
RDC
24
118
Photodet.
Amp.
40
PZT
100
48
64
116
Laser
HPF
Servo
PM
108
BPF
LPF
110
58
114
VCO
Mixer
112
106
30

FIG. 1-C

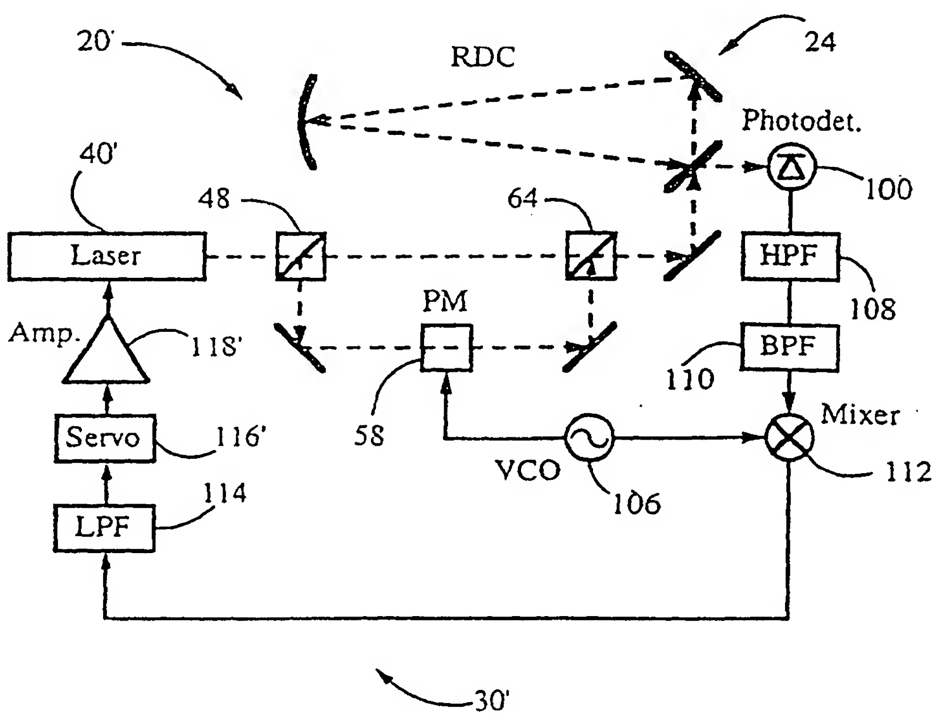

20'
RDC
24
Photodet.
40'
100
48
64
Laser
HPF
Amp.
PM
108
118'
BPF
110
Mixer
Servo
116'
58
VCO
112
106
114
LPF
30'

FIG. 2

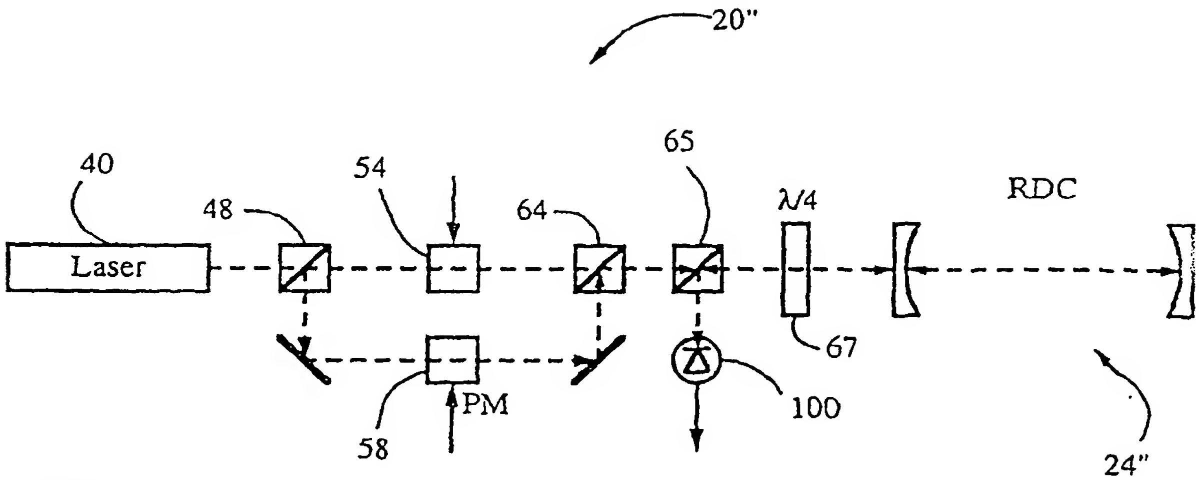

20"
40
Laser
48
54
64
65
λ/4
RDC
67
100
PM
58
24"

FIG. 3

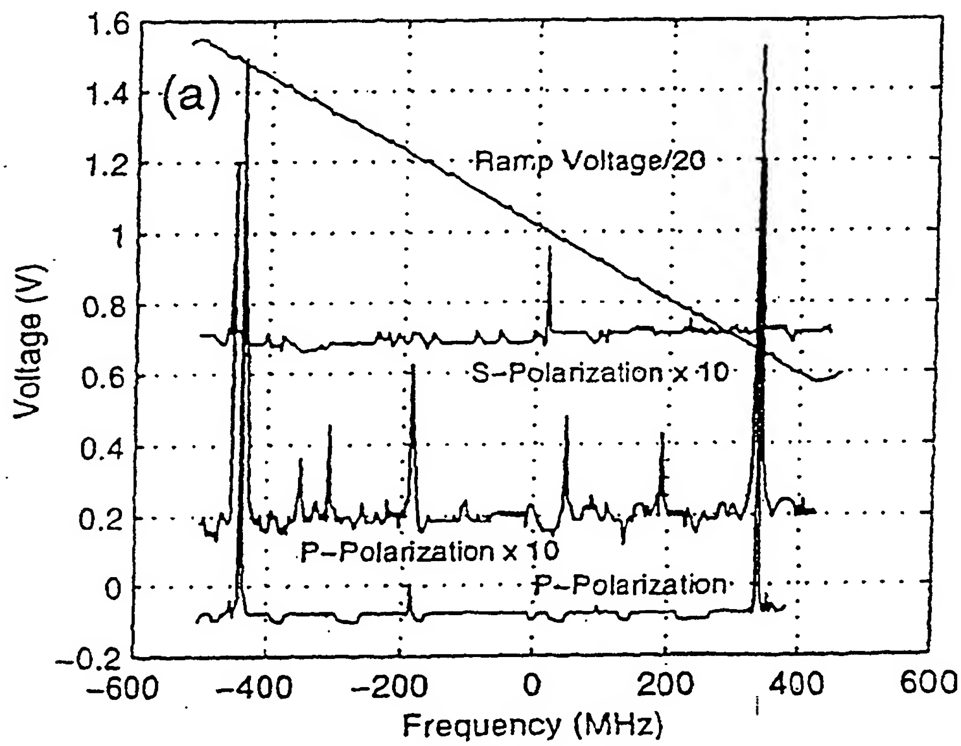
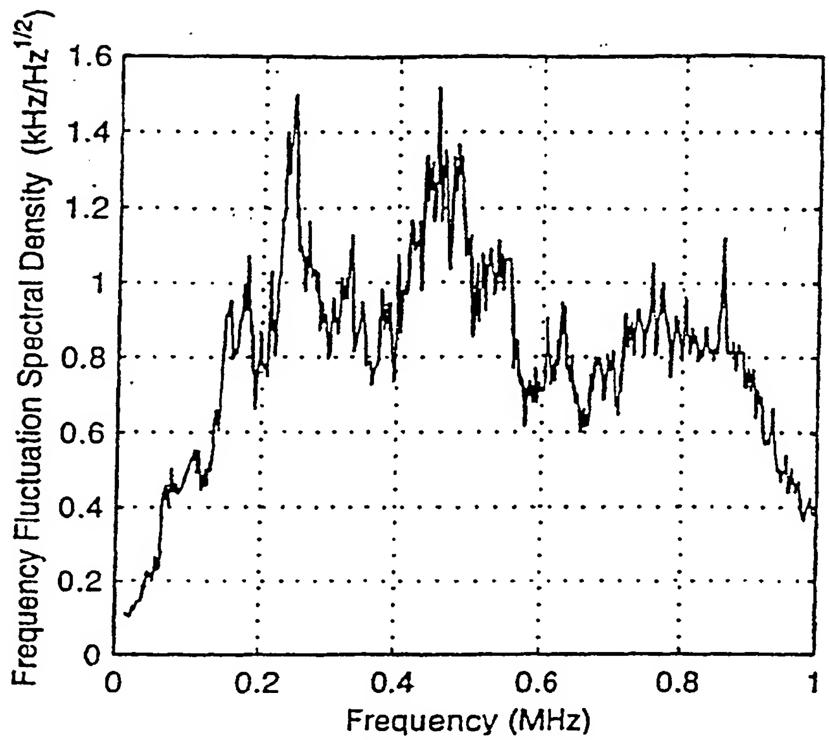

(a)
Ramp Voltage/20
S–Polarization x 10
P–Polarization x 10
P–Polarization
Voltage (V)
Frequency (MHz)
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
−0.2
−600
−400
−200
0
200
400
600

FIG. 4

FIG. 5


Frequency Fluctuation Spectral Density (kHz/Hz^1/2)
1.6
1.4
1.2
1
0.8
0.6
0.4
0.2
0
0
0.2
0.4
0.6
0.8
1
Frequency (MHz)

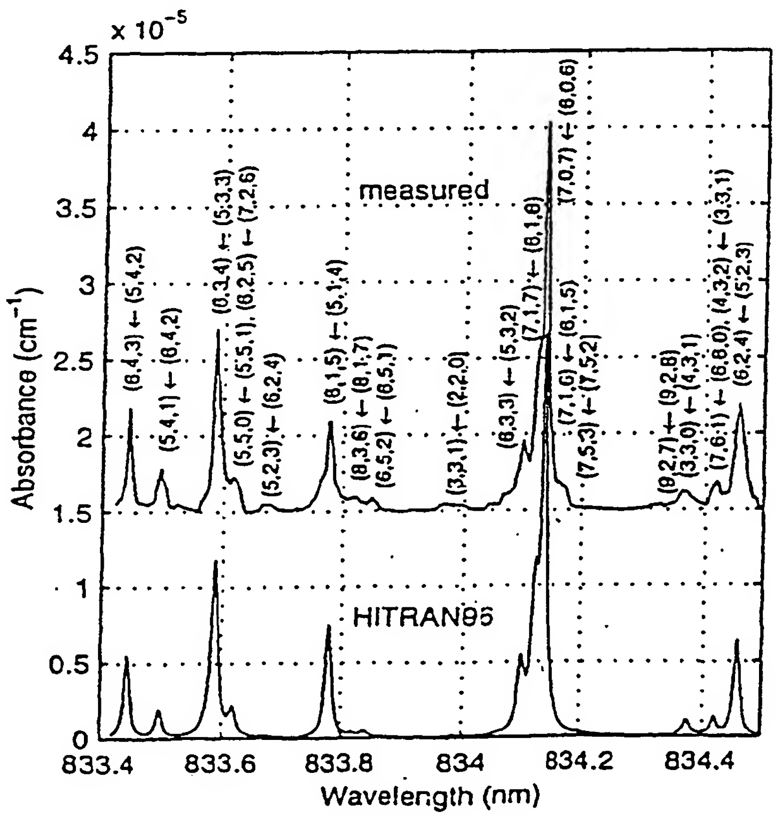

x 10^-5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
Absorbance (cm^-1)
measured
HITRAN96
(6,4,3) ← (5,4,2)
(5,4,1) ← (6,4,2)
(6,3,4) ← (5,3,3)
(5,5,0) ← (5,5,1), (6,2,5) ← (7,2,6)
(5,2,3) ← (6,2,4)
(6,1,5) ← (5,1,4)
(8,3,6) ← (8,1,7)
(6,5,2) ← (6,5,1)
(3,3,1) ← (2,2,0)
(6,3,3) ← (5,3,2)
(7,1,7) ← (6,1,6)
(7,0,7) ← (6,0,6)
(7,1,6) ← (6,1,5)
(7,5,3) ← (7,5,2)
(8,2,7) ← (9,2,8)
(3,3,0) ← (4,3,1)
(7,6,1) ← (6,6,0), (4,3,2) ← (3,3,1)
(6,2,4) ← (5,2,3)
833.4
833.6
833.8
834
834.2
834.4
Wavelength (nm)

FIG. 6

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 00 30 0294

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
X	PALDUS B A ET AL: "Cavity-locked ring-down spectroscopy" JOURNAL OF APPLIED PHYSICS, 15 APRIL 1998, AIP, USA, vol. 83, no. 8, pages 3991-3997, XP000910269 ISSN: 0021-8979 * figures 1,2A,2B,3A,4,6 * ---	1-14	G01N21/39 G01J3/433
A	PALDUS B A ET AL: "Advances in CW cavity ring-down spectroscopy" TECHNICAL DIGEST. SUMMARIES OF PAPERS PRESENTED AT THE INTERNATIONAL QUANTUM ELECTRONICS CONFERENCE. CONFERENCE EDITION. 1998 TECHNICAL DIGEST SERIES, VOL.7 (IEEE CAT. NO.98CH36236), TECHNICAL DIGEST SUMMARIES OF PAPERS PRESENTED AT THE INTERNATIONAL, page 79 XP000910287 1998, Washington, DC, USA, Opt. Soc. America, USA ISBN: 1-55752-541-2 * figure 1 * ---	1-14	
			TECHNICAL FIELDS SEARCHED (Int.Cl.7)
A	US 5 590 143 A (TAKARA HIDEHIKO ET AL) 31 December 1996 (1996-12-31) * claim 1 * ---	1-14	H05K G01N G01J
A	US 5 528 040 A (LEHMANN KEVIN K) 18 June 1996 (1996-06-18) * column 9; claims 1,14 * ---	1-14	
A	US 5 912 740 A (MARTIN JUERGEN ET AL) 15 June 1999 (1999-06-15) * claim 1; figure 1 * ---	1-14	
A	US 5 973 782 A (BOMSE DAVID S) 26 October 1999 (1999-10-26) * column 3 - column 4; claim 1; figure 2 * --- -/--	1-14	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
MUNICH	27 June 2000	Mason, W

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D : document cited in the application
L : document cited for other reasons

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EUROPEAN SEARCH REPORT

Application Number
EP 00 30 0294

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
A	US 5 636 035 A (WHITTAKER EDWARD A ET AL) 3 June 1997 (1997-06-03) * column 1-2; figure 1 * -----	1-14	

TECHNICAL FIELDS SEARCHED (Int.Cl.7)

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
MUNICH	27 June 2000	Mason, W

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X : particularly relevant if taken alone
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D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

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The members are as contained in the European Patent Office EDP file on
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27-06-2000

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US 5973782 A	26-10-1999	NONE	
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